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Compton DIV: Using a Compton-Based Gamma-Ray Imager for Design Information Verification of Uranium Enrichment Plants*

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A feasibility study has been performed to determine the potential usefulness of Compton imaging as a tool for design information verification (DIV) of uranium enrichment plants. Compton imaging is a method of gamma-ray imaging capable of imaging with a 360-degree field of view over a broad range of energies. These systems can image a room (with a time span on the order of one hour) and return a picture of the distribution and composition of radioactive material in that room. The effectiveness of Compton imaging depends on the sensitivity and resolution of the instrument as well the strength and energy of the radioactive material to be imaged. This study combined measurements and simulations to examine the specific issue of UF_6 gas flow in pipes, at various enrichment levels, as well as hold-up resulting from the accumulation of enriched material in those pipes. It was found that current generation imagers could image pipes carrying UF_6 in less than one hour at moderate to high enrichment. Pipes with low enriched gas would require more time. It was also found that hold-up was more amenable to this technique and could be imaged in gram quantities in a fraction of an hour. Another question arises regarding the ability to separately image two pipes spaced closely together. This depends on the capabilities of the instrument in question. These results are described in detail. In addition, suggestions are given as to how to develop Compton imaging as a tool for DIV.

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1.0 Introduction

A study was performed to examine the feasibility of Compton imaging for Design Information Verification (DIV) in a uranium-enrichment plant. Such a plant has a complex network of piping carrying UF_6 in gaseous form. Spectroscopic gamma-ray imaging offers the ability to determine the spatial distribution of radioactive isotopes that have a significant gamma-ray or hard X-ray signature. Since ^{235}U releases a significant amount of gamma radiation, there is the opportunity to determine which pipes are carrying enriched gas. Inspectors will use this information to look for diversion of material or excessive enrichment activity. Some of the questions that this technology could address include

- Which pipes have material flowing through them and which do not
- What is the isotopic composition of the material in each pipe
- Is there any hold up and if so, how much
- Has this flow or hold up changed from a previous inspection

Gamma-ray imaging as applied to this problem could be based on three different modalities: pinhole imager, coded aperture array, or Compton imaging. Each technique has its strengths and weaknesses. Gamma-ray images based on pinhole cameras are relatively simple to build and operate. However, their poor sensitivity immediately rules them out for these applications. Coded apertures cameras are a step up in complexity but offer much greater sensitivity. However, their limited field of view and poor performance in the presence of strong background radiation are significant drawbacks. Therefore, this study focused on the feasibility of Compton imaging cameras for the above-mentioned applications.

The challenges to such an effort are several. From an instrument point of view, these include

- *Energy resolution*: Excellent spectral resolution is needed to accurately determine the isotopic composition.
- *Imaging resolution*: Excellent imaging resolution is needed to separate one pipe from another.
- *Sensitivity*: Sufficient sensitivity is needed to make measurements within a reasonable time.
- *Practicality*: Reasonable cost, complexity and portability are needed to make the system practical.

2.0 Background

There are two basic methods to perform gamma-ray imaging. Collimator based imaging and Compton imaging. Collimator imaging (pinhole cameras, coded apertures, rotation modulating collimators etc.) rely on a tungsten or lead mask to cast a shadow of the source onto the detector. These systems have the advantage in that they are relatively simple and work well at low energies. The disadvantage is that the collimator blocks a large fraction of the signal, limiting sensitivity. In addition, they do not work well at higher energies where the gamma-rays begin to leak through the mask.

Compton imaging eliminates the need for a collimator. Instead, it relies on capturing the energy and position of each scatter site as the gamma ray interacts in the detector. Once this information is obtained, the famous Compton scatter equation can be used to determine the angle at which the source arrived at the detector.

$$\cos(\theta) = 1 - 511 \cdot \left(\frac{1}{E_2} - \frac{1}{E_{tot}} \right) \qquad \text{Compton scatter equation}$$

The advantage of Compton imaging is potentially high sensitivity compared to other techniques. The disadvantage is that it requires a more sophisticated instrument that can record the 3-D position of each interaction in the active detector material (Figure 1). Considerable work has been done developing such systems [2-4] and one such system was used for measurements described in this report (Figure 2).

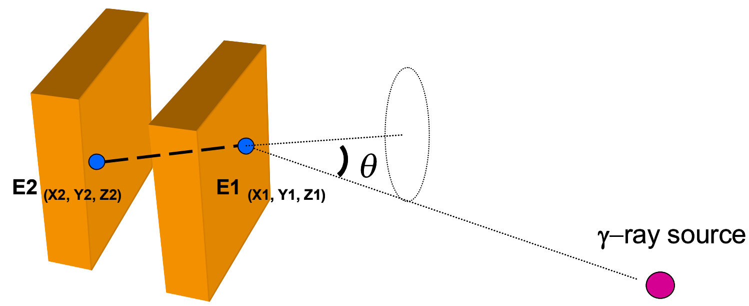

Figure 1. Schematic showing two detector planes of a Compton camera. As the gamma ray scatters through the planes, the energy and position of each interaction is recorded.

Figure 2. Compton imager known as CCI-1 on which many measurements were made. This instrument consists of a 38x38 channel germanium detector and a 32x32 channel silicon detector.

3.0 Methods

This report is based on a combination of measurements and Monte Carlo studies. The measurements came from an instrument called the Compact Compton Imager (CCI). This instrument is being developed separately. It represents the current state of the art in Compton imaging [2-4]. Measurements made on this instrument that are relevant to the present study include: extensive benchmarking to characterize the instrument; gamma-ray detection efficiency as a function of angle and energy; calibration; angular resolution studies. They also specifically included measurements of HEU in a quantity comparable to what is expected in holdup in a gaseous UF_6 pipe.

Simulations were performed in cases where relevant measurements were not possible. Specifically, this included the modeling of gaseous UF_6 in a pipe. This scenario was modeled using MCNPX and RADSRC [5,6]. These are described more below.

4.0 Determination of sensitivity of Compton imaging

The first step was to quantify the sensitivity of a Compton imager. Since the pressure of UF_6 in a pipe may be as little as 5 torr, the gamma-ray signal from it will be weak. Therefore, it is important to determine the minimum time needed to make an image.

For this measurement, a ^{22}Na point source (511 keV) was placed at 3 meters distance from the imager. The strength of the source was 13 micro-Curies and resulted in a flux of 27 counts per second at the detector. Figure 3 shows the resulting image which took 15 seconds to acquire using CCI and the current generation algorithms. The peak shown, near the center of the field of view, represents a minimum detection with approximately 3σ confidence. The image on the right side of figure 3 shows how the same image improves after several minutes of acquisition time.

This represents an imaging sensitivity of 0.36 micro-Curies at 511 keV at 1 meter in one minute. The sensitivity improves with longer acquisition times until it becomes limited by the environmental radiation background. This background is negligible for most measurements in a normal laboratory but could be considerable in other environments, such as an enrichment plant.

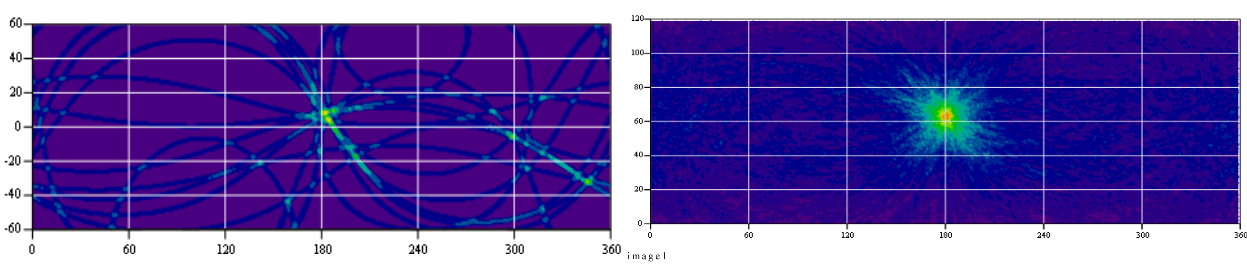

Figure 3. Two images from a 511 keV point source. The first image (left side) shows minimum (3σ) detection, requiring about 15 seconds. The second image shows a higher statistics image obtained after longer acquisition time. The horizontal axis represents 360° around the detector and the vertical axis represents +/- 60° above and below the detector. Thus these images show a nearly 4π field of view.

The quantitative analysis of imaging sensitivity, as in above, can be complicated. Imaging modalities, such as coded aperture and Compton imaging, have intrinsic artifacts. These artifacts result in correlated counting statistics that deviate from a strict Poisson distribution. However, in the case of low statistics (such as in determining minimum sensitivity) the deviation is small and can be safely ignored.

Finally, the sensitivity will be a function of energy. Higher energies will benefit from higher imaging resolution but poorer efficiency for stopping the gamma-ray. Lower energies will have poor resolution but high efficiency. To first order, these effects will cancel out. However, more detailed measurements of sensitivity versus energy would be useful in the future.

5.0 Modeling of pipes containing gaseous UF_6.

A Monte Carlo study was performed to answer questions about the feasibility of imaging a series of pipes containing gaseous UF_6 using a Compton camera. Two codes were used for this: MCNPX, a general purpose radiation transport code [5], and RADSRC, an LLNL-developed code to determine the gamma-ray emission from aged radioactive materials [6]. The goal was to determine if the signal generated would be sufficient for imaging. If so, how long would it take to get adequate statistics and at what distance.

The above measurement determined the sensitivity to a point source. In the case of pipes, the radioactive source will be distributed in a continuous fashion along one or more pipes. Currently, there is limited experience with imaging these types of sources. We instead relied on simulations and the assumption that a continuous source could be modeled as a superposition of point sources.

The pipes were assumed to be 3-mm thick stainless steel and 10 cm in diameter (4 inches). A "source pixel" was defined as a 10 x 10 cm region of pipe (Figure 4). The goal was to determine the minimum time and distance necessary to image each pixel.

Note that the imaging resolution may be better or worse than the selected pixel size of 10 cm x 10 cm as in this example. Ideally it is better. However, if it is worse, then blurring will occur between pixels in the image. A further complication arises because imaging resolution is energy dependent. However, the resolution can be improved by sacrificing field of view and moving the imager closer to the target. Thus, an optimum trade off must be found between resolution, field of view, and acquisition time.

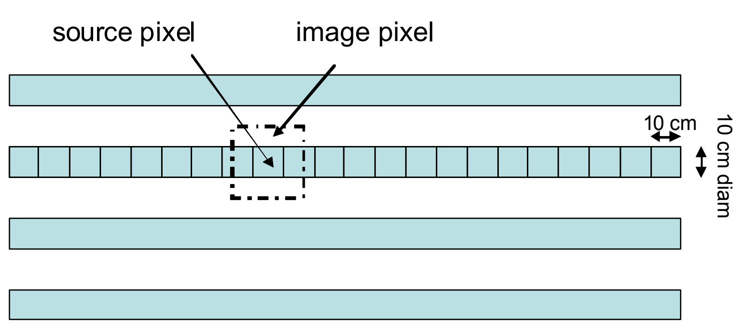

Figure 4. Schematic of several pipes showing the "source" pixel and "image" pixel.

The Monte Carlo study was carried out as a function of pressure and enrichment of the UF_6 gas. Pressures from 5 torr to 40 torr were examined as this is the range typically encountered in Uranium enrichment facilities. Three different isotopic compositions were considered, corresponding to enrichment levels of 5%, 20% and 92%. These are shown in table 1.

Table 1. Composition as modeled for three different enrichment levels

Enrichment	**5%**	**20%**	**92%**
^{235}U	0.0500	0.200	0.92
^{234}U	0.0005	0.002	0.009
^{238}U	0.9495	0.798	0.071

Note that the three cases contain only the isotopes of uranium but none of the daughter products. It is assumed that the gas has just undergone chemical separation in preparation for the centrifuges. However, any material that deposits on the pipe (hold-up) will decay and head towards equilibrium with its daughter products. This material will have a significantly different isotopic combination and resulting gamma-ray signature. This hold-up material will create a background that will compete with the imaging of the pure gas mixtures (discussed in the next section). Figure 5 shows the gamma-ray spectrum reaching the detector from the case of 92% enriched uranium hexafluoride gas.

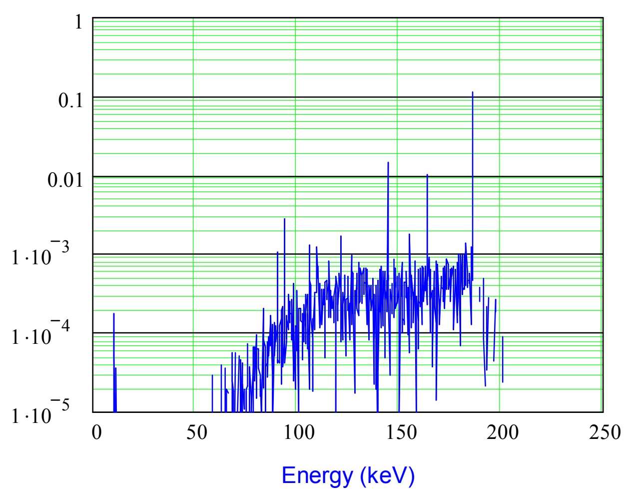

Figure 5. Simulated flux at the detector resulting from 92% enriched UF_6 at 5 torr. The detector has an active area of 100 cm^2 and was located at 1 meter from the pipe.

Similar spectra were made for 5 and 20% enrichments. With these spectra it is possible to estimate the counting time necessary to image pipes containing the three levels of enrichment. This is achieved by referring to section 4 where the minimum flux to perform and image was determined. As seen in the figure, the 186 keV line is the dominant line and only these events are considered for imaging. These results are shown in table 2.

Table 2. Estimated time for imaging UF_6 at 5 torr, assuming a 100 cm^2 detector at 1 meter

Enrichment	Estimated minimum imaging time
92%	0.23 hr
20%	1.13 hr
5%	4.53 hr

These results assume an instrument comparable to CCI, as described above. It is also assumed that the background due to hold-up or other sources is negligible.

Thus, the first conclusion is that it seems feasible to image pipes containing UF_6 at 5 torr. This assumes a detector comparable to existing technology located at 1 meter from the pipes. Higher pressure would result in correspondingly faster imaging times. However, the imaging time for low enrichment begins to be excessive. Furthermore, the spatial resolution at 186 keV suffers badly (as shown below). Therefore, it will be necessary to optimize the detector system and or develop imaging algorithms that are better suited for this energy range.

6.0 Imaging resolution

Another relevant set of measurements involves determining the imaging resolution as a function of energy. These measurements were made with a ^{152}Eu source which has a wide range of gamma-ray lines (Figure 6). These images represent the point spread function of the imager at each energy. The imaging resolution was measured at 1408, 344, 244 and 122 keV. At the higher energies the resolution is excellent, on the order of a degree or so. However, at around 244 keV and below, significant artifacts begin appear. In fact, at low energy the system loses the ability to image sources that are on-axis (around 0 degrees). This is a consequence of the physics of low energy scattering and the geometry of the detector. As discussed below, this is being addressed with an improved system design.

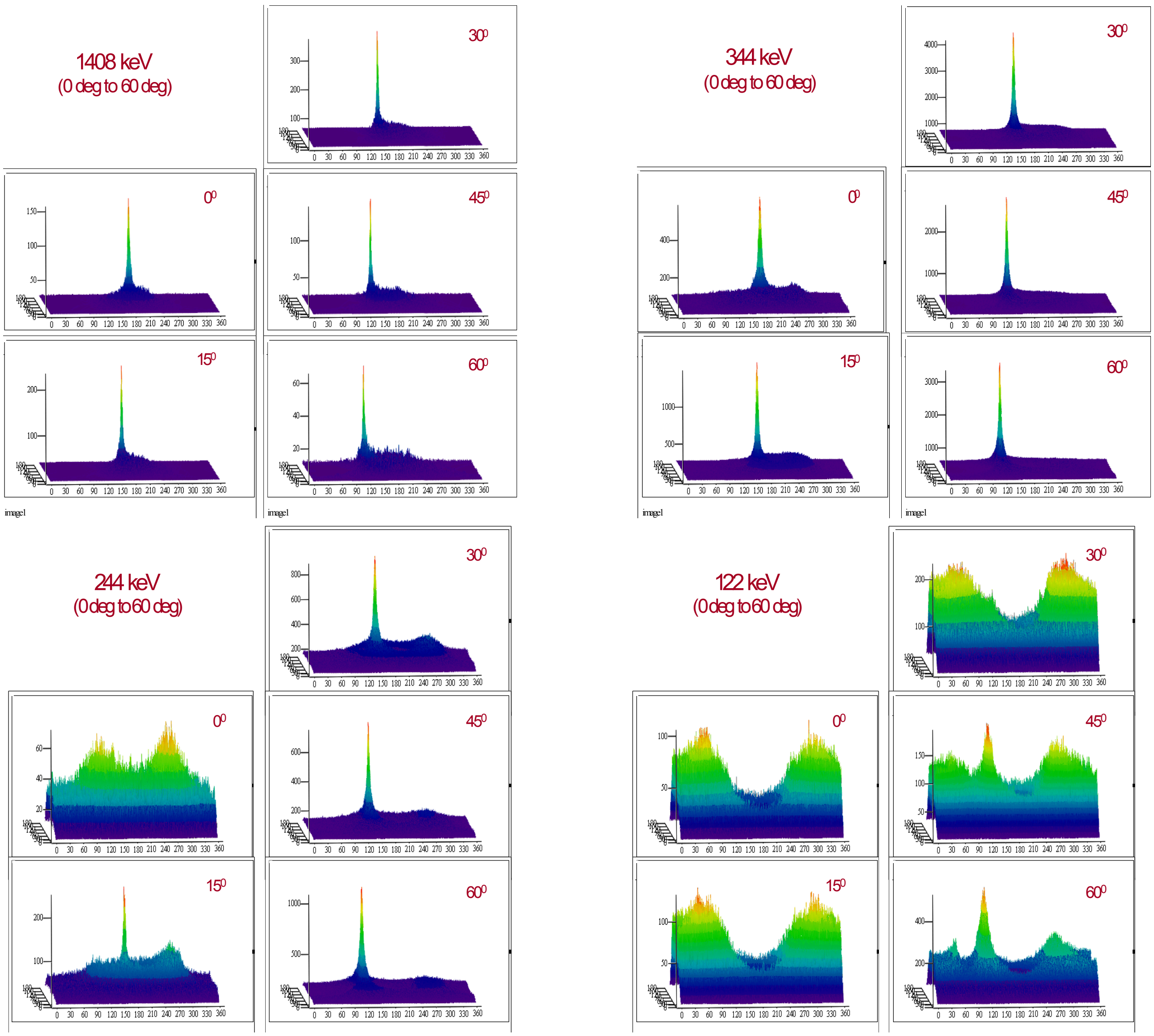

Figure 6. Point spread function as a function of angle for energies ranging from 122 keV to 1408 keV.

7.0 Measurement and simulation of hold up

In addition to the gaseous UF_6, there is a concern over hold-up material This material can take various forms. One form occurs when UF_6 mixes with water vapor to form UO_2F_2. This material precipitates out of the gas and plates onto the walls of the pipes. Another form comes from the natural decay of ^{238}U into Thorium. This also forms a solid on the walls. The hold-up material would initially have the same uranium isotopic composition as the gas, but over time would come to equilibrium with its daughter products.

Simulations were carried out to model this hold-up. Figure 7 shows the simulated flux resulting from approximately 1 gram of highly enriched uranium that has been deposited and come to equilibrium. Therefore, it includes not only the decays from the uranium isotopes but the daughter products as well. The same detector was assumed as in the previous measurements and simulations.

The dominant gamma-ray emission is still the 186 keV line. The flux is nearly three times as high from this simulated hold-up as from the gas. Such a quantity should be detectable in less than a minute, assuming it is localized.

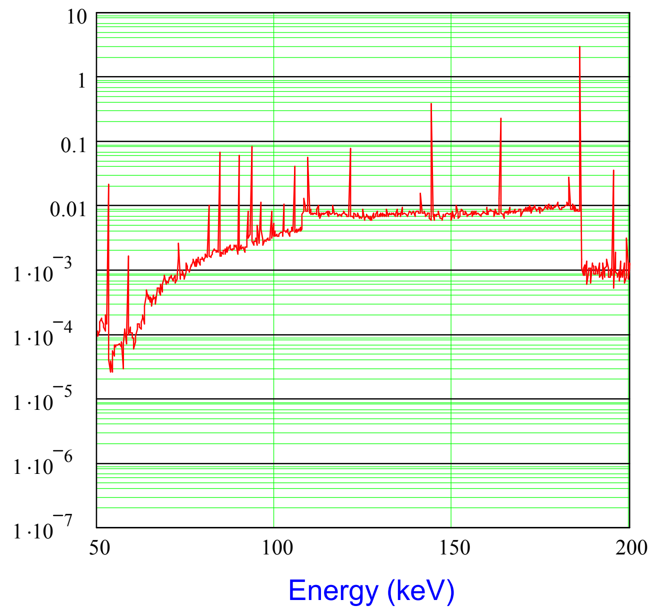

Figure 7. Gamma-ray flux resulting from approximately 1 gram of hold-up material resulting from 92% enriched UF_6.

In addition to the simulations, measurements were made using the same instrument, CCI, but for a separate application. This was done to verify the ability to image gram quantities of HEU [7]. For this measurement, a sample of 2.5 grams of HEU was imaged to determine the minimum time and distance required for detection. In addition, a sample of 400 grams of depleted uranium (DU) was used as a masking source. Figure 8 shows some of these results as a function of distance to the HEU. The DU was kept at a constant location and distance of 9 feet away from the detector. Each image was taken at 20 minutes acquisition time and a cut was made on the 186 keV photopeak events. At 4 feet, the HEU dominates the image and is clearly seen. At 5 to 6 feet, the HEU and DU both show up in the image. As the HEU continues to move farther away, it fades from the image, leaving only the DU visible.

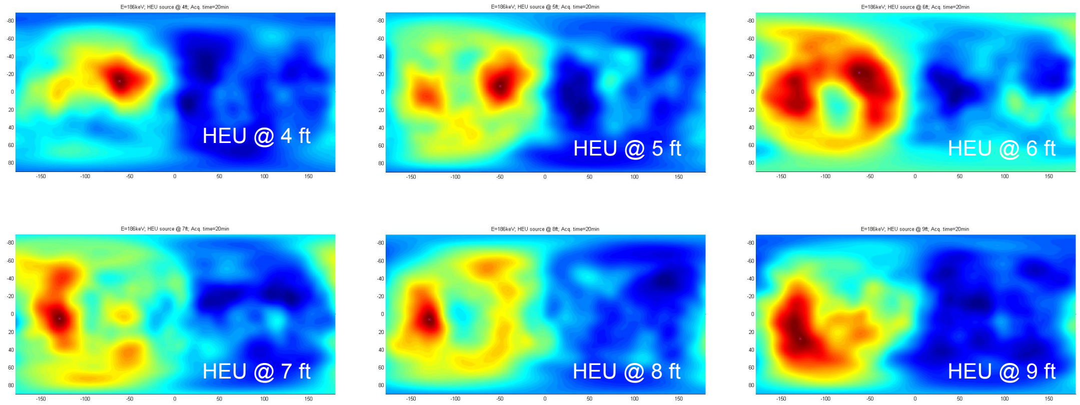

Figure 8. Two sources are seen in these images. A 2.5 gram HEU source and a 400 gram DU sources used as a making source. The HEU source was moved at different distances to determine the detection limits. The DU was kept at a constant 9 foot distance. The acquisition time of each image was 20 minutes. Each image was composed only of the 186 keV photopeak events.

This result is beneficial to the current study in that it shows that gram quantities of HEU can be easily imaged at close distances (few meters) on a time scale of tens of minutes. Furthermore, this can be done even in the presence of background or nuisance sources.

8.0 Issues not addressed in this report.

These measurements did not attempt to quantify imaging resolution, or to demonstrate imaging of distributed sources at low gamma-ray energies (below 200 keV). These will require dedicated measurements using HEU material in quantities of interest for monitoring enrichment plants. Also, measurements will have to be performed to quantify imaging resolution at 186keV. The results of these measurements can then be benchmarked against simulations.

9.0 Conclusions

A combination of simulations and measurements show that the signal emanating from pipes carrying highly enriched UF_6 is sufficiently strong that the pipes are amenable to imaging. Current generation technology could perform this imaging from a distance of a few meters in the times span of tens of minutes to a few hours. Furthermore, hold up material in gram quantities could be easily imaged within the time span of an hour.

However, the limitations of this technique are twofold. First, the signal from low enriched uranium will be weak and will take longer to image. Second, the spatial resolution of images at 186 keV, the main line of interest, will be poor with current generation technology. Therefore, the next section makes recommendations for a next-generation gamma-ray imager

10.0 Recommendations for a Next Generation Compton Imager

It is recommended that work be made towards a next-generation gamma-ray imager. This would combine the proven capability of existing Compton imaging technology but be optimized for safeguards applications. Specifically, such an imaging system would have the following properties.

1. **High energy resolution** for spectral specificity
2. **High imaging resolution** in the 186 keV region
3. **High efficiency** for detecting weak sources such as pipes with low enriched UF_6.
4. **Practical**: such as system must be small, portable and cost effective to be of use in real-world safeguards applications.

We are currently pursuing a new design of Compton imager to meet these goals. This instrument will employ developments in detector technology and electronics to yield higher resolution and efficiency while at the same time reducing the overall cost of a deployable system. A feasibility study has been completed and prototype development has begun.

11.0 References

1. The Imaging COMPTON Telescope COMPTEL on the Gamma Ray Observatory *Schönfelder V., Diehl R., Lichti G.G., Steinle H., Swanenburg B.N., Deerenberg A.J.M., Aarts H., Lockwood J., Webber W., Macri J., Ryan J., Simpson G., Taylor B.G., Bennett K., Snelling M.* IEEE Trans. on Nuclear Science, Vol. NS-31, No.1, 766-770 (1984)
2. M. Burks, D. Chivers, C. Cork, M. Cunningham, L. Fabris, D. Gunter, E. Hull et al., "Imaging Performance of the Si/Ge Hybrid Compton Imager", Proceedings of the IEEE Nuclear Science Symposium, October 2004.
3. *Vetter, K., Burks, M., Mihailescu, L., ``Gamma-ray imaging with position sensitive HPGe detectors'', NIM-A 525: 322-327 JUN 1 2004 (2004).*
4. *Mihailescu, L., Vetter, K., Burks, M., Hull, E., Craig, W., 'SPEIR: a Ge Compton camera', NIM-A, Vol. A570, pp. 89-100, (2007).*
5. Documentation of MCNPX found at http://mcnpx.lanl.gov/
6. Documentation of RADSRC found at http://nuclear.llnl.gov/
7. K. Vetter et al., "Technical Report – Part 2 Compact Compton Imager", Dec 2007